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Optimizing Cs_2LiYCl_6 for fast neutron spectroscopy

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ABSTRACT

Cs_2LiYCl_6 (CLYC) has generated recent interest as a thermal neutron detector due to its excellent n/γ-ray pulse-shape discrimination and energy resolution. Here, the capabilities of CLYC as a fast neutron detector and spectrometer are reported. A 1 in. × 1 in. CLYC detector was used to measure the response of mono-energetic neutrons over a range of 0.8–2.0 MeV produced via the $^{7}Li(p,n)$ reaction at the University of Massachusetts Lowell 5.5 MV Van de Graaff accelerator. A broad continuum from the $^{6}Li(n, \alpha)$ reaction was observed, as well as additional peaks below the thermal capture peak. Based on possible reactions in CLYC, the additional peaks are determined to be due to the $^{35}Cl(n,p)^{35}S$ reaction, with a *Q*-value of +615 keV, and corroborated in simulations using MCNPX. The average resolution of 9% for these peaks makes CLYC a promising candidate for a fast neutron spectrometer.

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1. Introduction

Measurement and spectroscopy of fast neutrons is a fundamental tool in many areas of research, including nuclear energy, stewardship science, nuclear safeguards, fusion plasma diagnostics, and fundamental nuclear physics. Fast neutron spectroscopy, however, continues to be a challenge due to limitations in existing detector technology. While time-of-flight techniques can produce accurate energy spectra, they are limited to situations in which a start signal, such as a correlated γ-ray or accelerator beam burst, is available. A compromise must also be made between improving resolution (with a longer flight path and thin detector) and increasing efficiency (with a shorter flight path and larger detector). Other spectrometers, such as proton recoil scintillators or ^{3}He proportional counters, involve unfolding measured spectra or the presence of a recoil continuum.

Over the past few years, Cs_2LiYCl_6 (CLYC) has been studied by several groups, including ours, as a thermal neutron and γ-ray detector. The optical properties of CLYC were first reported by the Delft group [1] in an effort to investigate chlorides as high light output scintillators. CLYC has since been used primarily as a thermal neutron detector through the $^{6}Li(n, \alpha)^{3}H$ reaction, which has a *Q*-value of 4.78 MeV. One of its primary advantages is its excellent n/γ discrimination capabilities, due to the large difference in decay times for pulses produced by electrons and those produced by the α-triton pair formed in neutron capture. Pulse-shape discrimination (PSD) with CLYC is typically done using the charge comparison method. Details on recent work describing PSD and thermal neutron detection with CLYC can be found in Refs. [2,3].

The thermal neutron peak resolution has been measured to be typically 2.5–3.1%. Therefore, though the $^{6}Li(n, \alpha)$ cross-section drops from 940 b for thermal neutrons to 0.24 b at 1 MeV [4], it may be possible to distinguish fast neutron induced reactions from thermal neutrons in a pulse height spectrum. For fast neutrons, the kinetic energy would be added to the *Q*-value of the reaction and shared between the α-particle and triton, depositing a total energy of E_n+Q in the crystal. In the past, this concept was applied to studying ^{6}LiI:Eu as a potential fast neutron spectrometer [5,6]. However, the resolution of the fast neutron peaks were found to be very broad (18%) at room temperature, and required that the crystals be cooled with liquid nitrogen. More recently, similar attempts were made using $LiBaF_3$:Ce [7], which again suffered from poor energy resolution (26% at 1.5 MeV).

Prompted by the excellent thermal neutron peak resolution in CLYC, a study of its potential fast neutron detection and spectroscopy capabilities have been carried out at the University of Massachusetts Lowell. Nearly mono-energetic neutron beams between 0.81 MeV and 2.02 MeV were produced at the 5.5 MV Van de Graaff via the $^{7}Li(p,n)^{7}Be$ reaction and detected with a 1 in. × 1 in. CLYC crystal. In analyzing the resulting neutron

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energy spectra a second reaction, identified as being due to ^{35}Cl, was found to contribute. While not originally anticipated, the ^{35}Cl(n,p)^{35}S reaction in CLYC appears to be promising for fast neutron spectroscopy.

2. Experimental setup

2.1. Neutron production

To produce mono-energetic neutrons, a thin natural Li target ($\approx$10 μm evaporated on a 0.54 mm tantalum backing) was mounted at the end of the Van de Graaff beamline and irradiated with a $\sim$15 μA DC proton beam, with energies of 3.0, 3.5, and 3.9 MeV. Mono-energetic neutrons are produced via the ^{7}Li(p,n)^{7}Be reaction, which has a threshold of 1.881 MeV. At these energies a second neutron group is produced from the ^{7}Li(p,n)^{7}Be* reaction, which has a threshold of 2.373 MeV. The energy of the first excited state of ^{7}Be is 431 keV. Therefore, the difference in energy between the first neutron group, coming from the ground state of ^{7}Be, and the second group from the first excited state was expected to be large enough that the two groups would be resolved. For a given proton energy, the angle between the beam direction and detector was also varied to obtain different neutron energies.

2.2. Detector setup

A 1 in. $\times$ 1 in. Cs_2LiYCl_6 crystal (provided by Radiation Monitoring Devices, Inc.) containing natural Li (7.42% ^{6}Li), was coupled to a Hamamatsu R6233-100 photomultiplier tube (PMT) and surrounded with a μ-metal magnetic shield and aluminum housing. To reduce the count rate of background thermal neutrons, the detector was entirely surrounded in a 3 mm thick layer of cadmium. The detector was positioned on a movable goniometer 54 cm from the target.

Because of the large ^{6}Li(n, α) cross-section at low energies, ranging from 940 b at thermal energies to 4.7 b at 1 keV with a $1/v$ behavior, neutrons scattered from materials in the target room was a concern. For a typical thermal neutron peak resolution of $\sim$5%, an energy of at least 240 keV would be required to distinguish a fast neutron from background. Since the cadmium cutoff is only 0.5 eV, there still remained a wide range of unresolvable background neutron energies that would fall within the so-called thermal peak.

Attempts were made to shield the detector with a thick annulus of paraffin and Li_2CO_3 to provide moderation and capture of low energy neutrons, but it was found that this only increased the number of counts in the thermal peak by 40%. This increase could be explained by the shield's moderation of fast neutrons from the beam down to energies that were both too high to be captured in the cadmium and too low to be resolved from the thermal peak.

Overall, it was determined that the best configuration was to have the detector surrounded only with cadmium. In future tests, more thorough investigations are planned for the source and shielding of background neutrons, some of which are known to originate from parts of the beamline prior to the target.

2.3. Data analysis

The PMT output signal was sent directly to a 1 GHz Lecroy Wavepro 7000 sampling oscilloscope for data collection. Raw signal traces were recorded within a 2 μs time window using a $-$20 mV threshold trigger, and analyzed off-line. At each energy, 80,000 signal traces were recorded using a sampling rate of 250 MHz. Though the scope could record traces at sampling rates of up to 1 GHz, higher sampling rates did not improve either the separation of n/γ pulses or energy resolution, but did increase data collection time by $\sim$20%.

In the off-line analysis, n/γ discrimination was performed using the charge comparison method, wherein two regions of the signal pulse are selected and integrated. The first region includes the sharp rise and peak, and the second includes the decaying portion of the signal. Since signals produced by neutrons and γ-rays are found to drastically differ in their decay time, the ratio of areas for these two integration windows differs depending on particle type. Two-dimensional plots of the ratio of window areas vs. total area were plotted. Neutron and γ-ray events are clearly distinguishable in these histograms and were sorted separately. Pulse-height spectra were produced by projecting the total pulse area of 2D neutron and γ-ray plots.

To calibrate and verify the position of the thermal peak, a data set was collected using a moderated PuBe source. The source was placed inside a container of paraffin large enough to thermalize approximately 75% of the original neutron spectrum and positioned 54 cm from the detector. A γ-ray calibration was carried out using the 1.274 MeV line from a ^{22}Na source. Applying this calibration to the thermal neutron peak places the 4.78 MeV ^{6}Li capture peak at an electron-equivalent energy of 3.5 MeV. The true energy of the thermal peak can then be obtained using a correction factor of 1.34. This method of calibration is not necessarily advisable though, as the relationship between γ-ray energy deposition and α-triton energy deposition may not be linear. Physically, this correction factor is indicative of the amount of light output quenching for the α-triton pair compared to electrons.

3. Results

3.1. n/γ Discrimination

Two signal traces representative of neutrons and γ-rays are shown in Fig. 1, where the two areas of integration, A_1 and A_2, used to calculate PSD parameters for each trace are indicated. The PSD parameter is defined as the ratio A_1/A_2. The widths of A_1 and A_2 were 128 ns and 160 ns, respectively, and were separated by 140 ns. These values were optimized by varying the width and position of each integration area for the best n/γ separation. It was found that the separation between γ-rays and ^{6}Li neutron events was largely insensitive to changes in the integration areas and always well separated. The basis by which the quality of n/γ

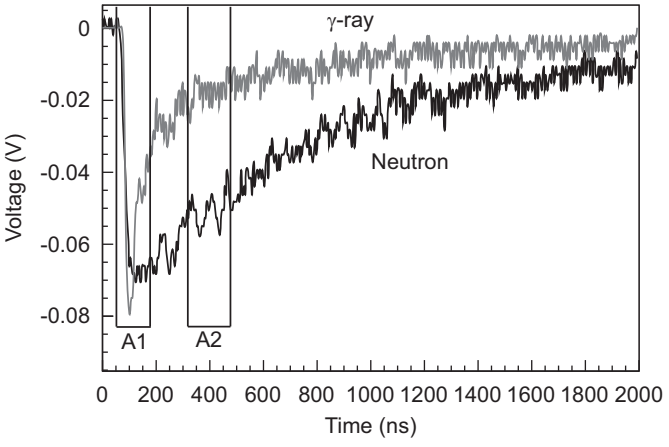

Fig. 1. PMT output signals for neutron and γ-ray. Integration areas A_1 and A_2 used to calculate PSD parameters, defined as A_1/A_2, are shown.